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An indicator to assess risks on water and air of pesticide spraying in crop fields

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Highlights:

- Stakeholders need tools to assess environmental risks of pesticide
- I-Phy3 assesses the risk on 3 environmental compartments (air / surface water / ground water)
- Knowledge of recent studies and expertise on pesticides are combined in I-Phy3
- I-Phy3 yielded better validation results than previous versions of the indicator
- A good compromise between ease of use and predictive capability is offered by I-Phy3

Abstract

Stakeholders involved in actions to reduce the use and the impacts on the environment or human health of pesticides need operational tools to assess crop protection strategies in regard to these impacts. I-Phy3 brings together all improvements introduced since the first version of the indicator to better meet user's needs and requirements of integrating processes. I-Phy3 was deeply modified to ensure its predictive quality. I-Phy 3 is structured in three levels of aggregation in form of hierarchical fuzzy decision trees designed with the CONTRA method. At the 1st level, five basic subindicators assess the risk of contamination (RC) for the different transfer pathways involved in surface water, ground water and atmosphere contamination: leaching, runoff, drainage, drift, volatilization. At the 2nd level, RC subindicators are aggregated with a toxicity variable (human or aquatic) in a risk indicator. At the 3rd level, the global indicator I-Phy3 results from the aggregation of three risk indicators for groundwater, surface waters and air. I-Phy3 yielded better validation results than its previous versions. This effort to assess the predictive quality of the indicator should be pursued and completed by a feasibility and utility test by end-users. A subindicator on risk of soil contamination is a gap which remains to fill.

43 Keywords: Indigo method; risk assessment; I-Phy3; pesticide transfer pathways; water
44 quality; air quality

45

46 1. Introduction

47 The incredible and securing rise of agriculture production since the end of the Second World
48 War, was favored partially by the widespread use of pesticides. Nevertheless, side effects on
49 the environment (Richardson, 1998) and in particular on water quality (Flury et al., 1995; Grung
50 et al., 2015; Lopez et al., 2015; Real et al., 2005), air (Hulin et al., 2021; Lichiheb et al., 2015)
51 and accumulation in soil (Silva et al., 2019; Tang and Maggi, 2021) have been observed. These
52 contaminations lead to exposure of non-target ecosystems as well as populations that may
53 have ecotoxicological and/or toxicological impacts. Consequently, regulations on pesticide use
54 have been continuously reinforced since the Council Directive 79/117/EEC with the Council
55 Directive EC 1107/2009 es and the Pesticide Package 2009/128/CE on sustainable use of
56 pesticides. This was amended by the Commission Directive (EU) 2019/782 introducing a
57 monitoring of risk by the HRI indicator (Methodology for calculating harmonized risk indicators
58 for pesticides under Directive 2009/128/EC, 2021) (European Commission. Statistical Office
59 of the European Union., 2021; (Lykogianni et al., 2021). In all cases, stakeholders involved in
60 actions to reduce the use and impacts of pesticides need operational tools to assess crop
61 protection strategies. The aim of such assessment may be to monitor and to report on the
62 current status of environmental compartments quality, to produce references for the good
63 management of crop protection and to work on innovative systems (Bockstaller et al., 2015).

64 The need for assessment tools dealing with pesticide issues has led to the development of
65 numerous indicators. The simplest ones rely on the supplied quantities, the Quantity of Active
66 Ingredients (QAI) or the Treatment Frequency Index (TFI) calculating the ratio of applied
67 pesticide to the registered rate (Hossard et al., 2017; Uthes et al., 2019). Although those
68 indicators have been developed to describe the evolution of pesticide use intensity, they are
69 often used as main indicators to address the environmental effects due to pesticide spraying
70 in environmental assessment method (Eckert et al., 2000; Vilain et al., 2008). Pesticide risk
71 indicators (Levitan, 2000) requiring complementary variables such as active ingredient
72 properties, crop management data and pedoclimatic variables are more elaborate and were
73 reviewed by several authors (Feola et al., 2011; Keichinger et al., 2013; Maud et al., 2001;
74 Reus et al., 2002). Some deserve more attention. While the Danish Pesticide Load (Kudsk et al.,
75 2018) is based on a quite simple scoring approach of pesticide properties,, other EPRIP2
76 (Trevisan et al., 2009),, POCER (Vercruysse and Steurbaut, 2002) or SYNOPS (Strassemeyer
77 and Gutsche, 2010), the indicators of the HAIR project (Kruijne et al., 2011)) rest on
78 quantitative equations addressing several factors (pesticide properties, soil climate, etc.). This
79 array of indicators may be explained by the context of use (purpose, environmental covered
80 addressed, scales, means, etc.) and the research of compromise between integration of
81 process and feasibility (Bockstaller et al., 2015) Among those indicators, the I-Phy indicator, in
82 its two versions, assessing risk for ground water, surface water and air, distinguished by its
83 original construction of a fuzzy decision tree combining qualitative and quantitative data. It was
84 developed for an assessment at the field level, first for arable cropping systems and later its
85 was adapted to viticulture (Thiollet-Scholtus and Bockstaller, 2015). Its aim was to help
86 advisers guide farmers in their choice of active ingredients and application methods at limit
87 pressure on the environment. In the validation study of Pierlot et al. (2017), I-Phy1 (first version,
88 van der Werf et Zimmer 1998) came out as one of the “best” indicators for transfer by leaching.
89 Lindahl et Bockstaller (2012) proposed an upgraded version assessing the pesticide transfer
90 by leaching to better take into account preferential transfer (I-Phy2), that was overlooked by I-
91 Phy1. However, its predictive quality was not improved.

92 Furthermore, assessments of transfer and toxicity of pesticides in I-Phy1 were combined in a
93 calculation of pesticide risk. Yet, stakeholders and advisers working directly with farmers
94 require a separate assessment of transfer to gain insight of water bodies contamination with
95 respect to the European rules for water bodies quality (DCE). This demand was motivated by
96 their need to gain knowledge on water quality in terms of pesticide contamination.
97 Ecotoxicological risk integrating transfer and toxicity remains a matter of interest when
98 stakeholders consider the impact of pesticide use on biocenosis. In both cases, the
99 implementation of assessment tools is aimed at helping them prioritize their actions.

100 Here is presented I-Phy3 which brings together all improvements introduced since the first
101 version of the indicator to better meet users' needs and requirements of integrating processes
102 to ensure its predictive quality. Based on the knowledge gained on pesticide transfers and risks
103 for the last 20 years, the indicator was restructured to separate transfer assessment from risks
104 on living organisms assessment, new input variables were introduced and most algorithms
105 deeply revised. This article presents the new calculation algorithms for arable crops as well as
106 results of the indicator evaluation for its predictive quality. Besides the design of the indicator
107 and validation results, possibilities for implementing the indicator are discussed.
108

109 **2. Material and methods**

110 The design of I-Phy3 started with the selection of input variables (section 2.1). Those variables
111 were aggregated into several subindicators and then in a global indicator I-Phy3. The
112 methodology is described in section 2.2 and the structure of the indicator in section 2.3. The
113 predictive quality of the indicator was then assessed by comparison of indicator outputs
114 calculated on sites with measured values of water or air contamination. Section 2.4 presents
115 the study sites used to collect measurements of water (EQUIPE project) or air (Repp'Air
116 project) contaminations while section 2.5 presents the methodology to assess predictive
117 quality of the indicator.

118

119 **2.1. Selection of input variables**

120 Input variables were selected from a literature review, starting from the work of van der Werf
121 (1996) and on expertise based on experimental data (e.g. see section 2.3.2 for contamination
122 of groundwater).

123 For pesticides properties, we used the revised Pesticide Properties Database PPDB which is
124 updated since more than 20 years with more reliable value available for each active ingredient
125 (a.i.) and used in many risk assessment studies (Lewis et al., 2016). PPDB can be browsed
126 on a website (PPDB : Pesticide Properties Database, 2020) or purchased in form of an Access
127 database. Values of pesticide properties were extracted from the latter (version
128 20/05/11).(Tomlin et al., 1995) For the DT50, the value proposed for laboratory rather than the
129 "typical value" proposed by the database resulting from an expert work was selected. This
130 choice resulted also from the calibration work with monitoring data in the Rhine plain (ERMES
131 2017, Koller et al. 2015) for four active ingredients (see section 2.3.3). For some few active
132 ingredients, values as listed in Supplementary Materials S1 were modified. For some few other
133 a.i., values were taken from other sources because of missing value in PPDB. Overall, 498
134 active ingredients were integrated in the database.
135

136 **2.2. Design of indicators**

137 The indicator I-Phy3 as well as all subindicators are calculated for a single active ingredient
138 and then aggregated for a spraying program. They are expressed on a continuous
139 performance scale between 0 (highest risk) and 10 (lowest risk), which allows an operational
140 user of the indicators by non-scientific users. (Bockstaller et al., 2008). (Craheix et al., This
141 scale can be easily inverted between 0 (lowest risk) and 10 (highest risk) for a use of I-Phy3
142 for specific risk assessment for which high values of indicators are usually associated to high
143 risk".

144 Fuzzy decision trees were implemented to design the indicator I-Phy and its different
145 subindicators like in the first version of the indicator (Roussel et al., 2000; van der Werf and
146 Zimmer, 1998). Fuzzy decision trees present several advantages (Bockstaller et al., 2017):
147 they rely on linguistic "if then" rules with a transparency, at least semantic through linguistic
148 rules that are easy to understand for non-specialists. They cope with qualitative as well as
149 quantitative heterogeneous information. Furthermore, combining decision trees with fuzzy
150 logic makes it possible to mitigate threshold effects linked to the linguistic "if then" rules when
151 they are Boolean, i.e., consisting of two alternatives yes/no. Fuzzy logic introduces fuzzy
152 subsets to deal with the whole set of intermediate cases.

153 As shown in the simplified example , (Supplementary Materials S2), I-Phy3 and each
154 subindicator require i) fuzzy subsets for each input variable with threshold values, ii)
155 membership functions to calculate membership degree of each variable to the favourable (low
156 risk) or unfavourable (high risk) subset, iii) decision trees. More details for fuzzy subsets and
157 membership functions are given in Supplementary Materials S3, while decision trees are
158 presented in section 2.3. Most decision trees were designed with the CONTRA method which
159 supports the design of fuzzy decision tree in a transparent way(Bockstaller et al., 2017) were
160 calibrated with a model (see section 2.3.3) or based on one variable (see sections 2.3.5 and
161 2.3.6). In this latter case, the membership degree to the favourable subset (expressed between
162 0 and 1) is transformed into an indicator score by multiplying the former by 10.

163

164 **2.3. Description of the indicator**

165 **2.3.1. Overview of the indicator structure and calculation**

166

167 As shown on Figure 1, I-Phy3 is structured in three levels of aggregation. At the 3rd level, the
168 global indicator I-Phy3 results from the aggregation of three risk indicators tackling three
169 environmental compartments like in version 1 (van der Werf and Zimmer, 1998): 1)
170 groundwater, 2) surface water and 3) air. At the 2nd level, each of this indicator of risk consists
171 in the aggregation of one or two sub indicators assessing the risk of contamination (RC) via
172 specific transfer pathways, with a toxicity variable, Admissible Daily Intake (ADI) or Aquatox
173 (highest toxicity level between fish, daphnia or alqua, for the human health and ecotoxicological
174 impacts respectively, see van der Werf and Zimmer (1998)). At the 1st level, five basic
175 subindicators assess the risk of contamination (RC) namely, a) the groundwater contamination
176 by pesticide leaching ($RC_{gw_{lea}}$), b) surface water contamination by pesticide runoff or drainage
177 transfer ($RC_{sw_{rd}}$). Drainage transfer is characterized by an initial vertical transfer, *i.e.* leaching,
178 to an impermeable layer where till drains have been set to evacuate rapidly water excess. This
179 requires a specific calculation (see 3.1.4). c) surface water contamination by pesticide drift
180 ($RC_{sw_{dr}}$); d) air contamination by pesticide volatilization ($RC_{air_{vol}}$) and e) air contamination by
181 pesticide drift ($RC_{air_{dr}}$).

182 The five risks of contamination are calculated for 1 kg of active ingredient (a.i.) and modulated
183 by the actual application rate which results from the calculation of a non-intercepted rate of a.i.
184 by the treated crop (see section 2.3.8): each of these five subindicators is transformed in a risk
185 between 0 (no) and 1 (high) and weighted by the percentage of sprayed area within the field.
186 It is then retransformed in a value of performance according to Equation 1. It was assumed
187 that there is a proportional relation between sprayed area and level of contamination (Melland
188 et al., 2016):

189

$$190 \quad RC_{final} = 10 \left(1 - \left(1 - \frac{RC_{100\%}}{10} \right) \right) \frac{\%Area}{100} \quad (\text{Equation 1})$$

191 Where:

192 RC_{final} : final result for a risk of contamination (see Figure 1)

193 $RC_{100\%}$: result for a risk of contamination calculated for 100% of sprayed area

194 %area: percentage of sprayed area within a field.

195 I-Phy and each subindicator result from the aggregation based on a fuzzy decision tree.

196

197

198 **2.3.2. Groundwater contamination ($RC_{GW_{lea}}$)**

199 Two variables of previous versions were kept: the GUS and the leaching potential for which a
200 new calculation method was proposed (Supplementary Materials S4.1.). A variable identified
201 as playing an important role was added: the water status of the soil when pesticides are applied
202 (Pierlot et al., 2017) depending on climate and soil conditions. This variable was assessed
203 through the application period (Supplementary Materials S4.2). Finally, solubility as a fourth
204 variable was added. This property was identified as explaining of the discrepancy between the
205 low transfer risk calculated by previous indicator version and the alarming contamination level
206 of groundwater in Rhine Plain for the nicosulfuron active ingredient (ERMES, 2017).
207 Nicosulfuron is characterized by a very high solubility in water (7500 mg.L⁻¹). The role of
208 solubility in pesticide leaching is confirmed by literature (Elliott et al., 2000). While pesticide
209 properties are directly retrieved from data bases, the two other variables have to be calculated
210 (Supplementary Materials 4.1 and 4.2).

211 No prominent weight was assigned to the GUS, like the version 1, regarding the aggregation
212 of the four input variables in a fuzzy decision tree (Table 1). This is justified by the fact that in
213 situations of preferential transfer, even active ingredient with favourable GUS like glyphosate
214 can be leached (Vereecken, 2005). We selected weights and modified some decision rules
215 proposed automatically by CONTRA as made possible by this method, to adjust indicators
216 outputs to results of groundwater monitoring of the Rhine Plain for four active ingredients
217 among the most used pesticides for the main crop of the region, maize' (dmat-p, mesotrion,
218 nicosulfuron, s-metolachlor), (ERMES, 2017; Koller et al., 2015). Table 1 depicts the structure
219 of the indicator.

220

221 **2.3.3. Surface water contamination by drainage/runoff** 222 **($RC_{sw_{dr}}$)**

223 The calculation of this subindicator, coming from the work of Wohlfart (2008) is based on the
224 runoff potential and a availability variable that is more elaborated than the use of the single
225 DT50 in the first version (van der Werf and Zimmer 1998). The runoff potential depends on
226 slope, on soil properties, texture, crusting sensitivity, hydromorphy, and also on management,
227 tillage and implantation of buffers strip (Supplementary Materials S5). The availability variable
228 is inspired from quantitative indicators like Synops (Strassemeyer and Gutsche, 2010) and
229 Eprisp2 (Trevisan et al., 2009). Equation 2 shows the calculation of the availability variable:

$$230 \quad \mathbf{Avai} = r \cdot e^{-Ln(2)/(DT50 \cdot t)} \quad (\text{Equation 2})$$

231 Where:

232 Avai : availability of active ingredient

233 DT50 : soil half-life of active ingredient (days)

234 t : time in days between date of spraying and date of next runoff event with a default value of
235 3 for a worst-case situation

236 r: A reduction coefficient assessing the reduction pesticide amount for runoff when the active
237 ingredient is incorporated (Mickelson et al., 2001). Considering the results of these authors,
238 we proposed a default value of 0.5 when incorporation and 1 when no incorporation.

239

240 Table 2 depicts the fuzzy decision tree aggregating those two variables. Outputs of the decision
241 trees were calibrated with help of the PRZMv3.12 model (Carsel et al., 1986) for two rates
242 (Wohlfahrt, 2008) and interpolated for 1 kg. Input variables belong either to the favourable set
243 (F) or to unfavourable set (U).

244

245

246 **2.3.4. Particular case of drained plot**

247 It has been clearly demonstrated that drained fields show a high risk of rapid transfer of
248 pesticide to adjacent surface water bodies through subsurface pipes (Brown and van Beinum,
249 2009). Although the initial transfer process consists in a preferential vertical transfer, final
250 impacted compartment is surface water and not groundwater. This is supported by the
251 variables playing a role in the determinism of pesticide loss in drained field (Brown and van
252 Beinum, 2009), Kd (non-normalized KOC) and DT50 which are both aggregated in the GUS
253 variable main variable for the groundwater subindicator (see section 2.3.1.). To cope with this
254 specificity, was assumed that in drained field, pesticides are mainly transferable to surface
255 water and not groundwater although the transfer process is based initially on vertical leaching
256 like for RCgw_{lea} (see section 2.3.1.) when there is no higher risk by runoff.

257 Therefore, the following specific decision rules was introduced:

258

259 **If the field is drained then RCsw_{dr} = MIN(RCsw_{dr} , RCgw_{lea})**

260 **If the field is drained and RCsw_{dr} = RCgw_{lea} then RCgw_{lea} =10**
261 (to avoid double counting of risk)

262 Where:

263 $RC_{sw_{dr}}$: contamination risk for surface water through drainage or runoff (see section 2.3.3)

264 $RC_{gw_{lea}}$: contamination risk for groundwater through leaching (see section 2.3.2)

265

266 **2.3.5. Surface water contamination by drift ($RC_{sw_{dr}}$)**

267 While in the previous version of the indicator, coefficients were taken from a table resulting
268 from a collection of expert judgment and measured values, the equations of Trevisan et al.
269 (2009) were used here to calculate spray drift. This spray drift potential is divided by 5 when
270 anti-drift nozzles are used. These spray drift values are transformed in a score between 0 and
271 10 as described in Supplementary Materials S6.

272

273 **2.3.6. Air contamination by volatilization ($RC_{air_{vol}}$)**

274 Like for the subindicator *spray drift to surface water* (see previous section), the decision tree
275 used in previous version of the indicator was replaced by quantitative equations calculating
276 pesticide volatilization (expressed in $\mu\text{g}/\text{m}^2/\text{hr}$) in function of pesticide properties (Woodrow et
277 al., 1997). We added some abatement factors taking into account effect of soil components
278 (mulch, etc.), pesticide properties (penetration ability), and field edge (presence of hedges or
279 trees reducing transfer to outside of field) which is expected to contribute to decrease the
280 transfer risk. The calculation is given by Equation 3 and more details are given in
281 Supplementary Materials S7.

282

$$284 \quad Vol_{tot} = (1 - c_{edge}) \cdot ((1 - c_{form}) \cdot (1 - ic) \cdot Vol_{sol} + (1 - c_{prod}) \cdot ic \cdot Vol_{plant})$$

283 (Equation 3)

285

286 Where:

287 Vol_{tot} : total volatilization (expressed in $\mu\text{g}/\text{m}^2/\text{hr}$)

288 Vol_{sol} : volatilization from soil (expressed in $\mu\text{g}/\text{m}^2/\text{hr}$), calculated according to Woodrow et al.
289 (1997): $\text{Ln}Vol_{sol} = 28.335 + 1.6158 \cdot \text{Ln}(Pv / (KOC \cdot Sol))$ with: Pv: pressure vapor (Pa), KOC: soil
290 adsorption coefficient ($\text{mg} \cdot \text{L}^{-1}$), Sol: water solubility ($\text{mg} \cdot \text{L}^{-1}$)

291 Vol_{plant} : volatilization from plant (expressed in $\mu\text{g}/\text{m}^2/\text{hr}$), calculated according to (Woodrow et
292 al., 1997): $\text{Ln}Vol_{plant} = 11.779 + 0.85543 \cdot \text{Ln}(Pv)$ with: Pv; pressure vapor (Pa)

293 ic: interception of pesticide by crop (see 3.1.8 and Supplementary Materials S8.1)

294 c_{edge} : abatement coefficient (between 0 and 1) due to field edge, more precisely to the presence
295 of a hedge reducing pesticide transfer to outside of field. Four variables are used to assess it:
296 type of plant (persistent or deciduous), hedge density (number of field sides with a hedge and
297 porosity of hedge), hedge height, spraying month (to assess the presence of leaves or not)

298 c_{form} : abatement coefficient (between 0 and 1) due to the product formulation

299 c_{prod} : abatement coefficient (between 0 and 1) due to penetration ability of the product in plant
300 which limits pesticide volatilization and is assessed with pesticide mechanism of action, the
301 octanol water coefficient (LogKow) and the use of an adjuvant to facilitate penetration.

302

303

2.3.7. Air contamination by drift ($RC_{air_{dr}}$)

304 This risk of air contamination by spray drift was not covered by the original version of I-Phy
305 (van der Werf and Zimmer, 1998) but was added to the version for wine growing activity (with
306 a simplified assessment including only the type of sprayer. A more elaborated decision tree
307 taking into account additional relevant variables like speed sprayer, the sprayer height, the use
308 of antidrift nozzle and the air pressure (Pressure) were included in this new version of the
309 indicator according to the study of Bahrouni, Sinfort, et Hamza (2010) (Table 3). The CONTRA
310 method (was used to aggregate the five input variables in a fuzzy decision tree. This indicator
311 is weighted by an abatement coefficient, c_{edge} like for $RC_{air_{vol}}$ (see Equation 3 and
312 Supplementary Materials 7.2).

313

314

315

2.3.8. Integration of the a.i. application rate effect

316 In I-Phy3, the method proposed by Lindahl and Bockstaller (2012) was used to integrate the
317 effect of the a.i. application rate in the calculation of each contamination subindicator. They
318 proposed to calculate an effective dose of a.i. available for transfer by weighting the initial dose
319 by an interception rate as shown in equation 4:

$$320 \text{Dose}_{\text{eff}} = (1-ic).\text{Dose}_{\text{ini}} \text{ (Equation 4)}$$

321

322 Dose_{eff} : effective rate of pesticide

323 ic: interception coefficient (see Supplementary Materials S98.1)

324 Dose_{ini} : initial rate of pesticide

325 They designed an algorithm which makes possible to reduce (if the dose is higher than 1 kg.ha⁻¹)
326 or increase (if the rate is lower than 1 kg.ha⁻¹) the indicator value obtained for 1 kg/ha⁻¹ a.i., in
327 function of this effective dose. More details on the determination of the interception rate and
328 the calculation of the indicator in function of the effective dose are given in Supplementary
329 Materials S 8.

330

331

2.3.9 . Risk by compartment

332 Three subindicators of environmental risk, for ground water ($I\text{-Phy}_{gw}$), surface water ($I\text{-Phy}_{sw}$),
333 and air ($I\text{-Phy}_{air}$) result from the aggregation of the five subindicators of contamination risk (RC)
334 with toxicity variables (see Figure 1). For groundwater, $RC_{gw_{res}}$ is aggregated with the daily
335 admissible intake (ADI) because this source is for human water supply. For surface water, two
336 subindicators, $RC_{sw_{res}}$ and $RC_{sw_{a}}$ are aggregated with a toxicity variable based on the highest
337 toxicity between Aquatox tackling toxicity for aquatic organisms and ADI for human toxicity
338 (Roussel et al., 2000). Aquatox results from the highest toxicity for fish, daphnia and algae
339 (van der Werf and Zimmer, 1998). For air, two subindicators, $RC_{air_{vol}}$ and $RC_{air_{dr}}$ are aggregated
340 with ADI for human toxicity.

341

342 Regarding aggregation rules, a weight of 60 % is given to the contamination risk and 40 % to
343 toxicity for groundwater (Supplementary Materials S9). For surface water and air, 30 % is given
344 to each contamination risk and again 40 % to the toxicity with a small modification in the second
345 case (Table 4). We considered that a situation with a high risk of contamination is more
346 undesirable than a situation of toxic a.i. without any risk of contamination. This may be justified
347 by water potability threshold of $0.1 \mu\text{g}^{-1}$ applied to all a.i. which leads water manager to focus
348 on water contamination. Furthermore, uncertainty exists on actual toxicity of a.i. which is not
349 well assessed by regulation tests (Centner, 2021) so that a situation with apparently no toxicity
350 may present a risk for human health. Similar decision rules to those for I-Phy_{sw} are set to I-Phy_{air}.

351

352

2.3.10. Final aggregated indicator (I-Phy)

353 The three subindicators of pesticide risk per compartment, ground (I-Phy_{gw})- and surface water
354 (I-Phy_{sw}), air (I-Phy_{air}) are aggregated with the same weight of 33% given to each compartment
355 . However, to limit compensation, the score was reduced to 6 when there was a risk totally
356 unfavourable for one compartment (I-Phy_{gw} or I-Phy_{sw} or I-Phy_{air}=0) and to 2 when two
357 compartments were concerned by a totally unfavourable risk (Table 5). We consider that if
358 there is a risk maximal for one compartment, the value should be clearly under 7. This value 7
359 is a reference value expressing an acceptable risk for the environment used for the set of
360 indicators of the INDIGO method to which belongs I-Phy (Bockstaller et al., 1997).

361

362

2.3.11. Implementation of the indicator

363 Calculations of the indicator are run on an Excel Sheet with one sheet in which all data on input
364 variables are entered. Each line corresponds to a calculation for one active ingredient. Stable
365 data (e.g. field characteristics) have to be copied from line to line. Users have access to all the
366 detail of calculations with results expressed with a color code (see Figure 3).

367

368

2.4. Study sites

369

370 Measured data of environmental compartment contamination (groundwater, surface water, air)
371 from several study sites (Figure 2) were used and compared with outputs of I-Phy3 for
372 validation. The sites of the EQUIPE project (see 2.3.1) provided data on the transfer by
373 drainage or runoff to surface water and vertical transfer by leaching. For the transfer to air, the
374 data of the project Repp'Air was used. For each treatment, the I-Phy 3 indicator was calculated
375 with the help of an Excel sheet calculator with the aim of comparing the results with
376 measurement data.

377

378

2.4.1. Sites of the Equipe project

379 The EQUIPE (2014-2017) project aimed to assess the predictive quality of pesticide indicators
380 addressing transfers to surface and ground water. To do so, outputs of 26 indicators (among
381 them Synops, Eprisp, I-Phy1, I-Phy2...) and a mechanistic model (MACRO) were compared to
382 measure pesticide transfers at plot levels at four sites with different climate and soil conditions,
383 and transfer pathways (Pierlot et al., 2017). The complete description of the 3 sites and the
384 indicators and model tested is detailed in Supplementary Materials S10) The Jailleire

385 experimental station, located in the Pays de la Loire region (France), is under the influence of
386 an oceanic climate, with a brown hydromorphic clay-textured soil, resulting from alterite shale.
387 This experimentation site consists of 10 agricultural plots of 0.5 to 1 ha each, where water from
388 drainage and runoff (saturation overland flow) are collected separately; ii) The experimental
389 station of the Magneraud, located in the Nouvelle-Aquitaine region (France), is also under the
390 influence of oceanic climate and is composed mainly of clayed and silty limestone soils
391 developed on sand-stone strata characterized by alternating layers of hard limestone and marl.
392 This site is made up of 14 lysimetric plots of 1 m² surface, with no vertical walls and no soil
393 shuffle; and iii) the Geispitzen experimental station is located in the hills of the lower Sundgau
394 district (Alsace region, France) and has an attenuated oceanic climate. The hills are covered
395 with loess-derived soils of silt loam texture overlying Oligocene molasses and marls. A sloping
396 field (5%) of about 9 ha was divided into 3 bordered fields with measuring flumes and automatic
397 water samplers at the down slope borders just upslope of a ditch drainage catchment runoff.

398

399 **2.342. Sites of the Repp’Air project**

400 The 7 measurement sites selected for the Repp’Air project came from historical sites monitored
401 by regional Association of Air Quality Survey in association with Chambers of Agriculture in
402 order to have different agricultural systems: arable crops, vineyards, arboriculture, mixed
403 cropping-livestock, and “mixed” sites with different types of crops. Farm practice surveys were
404 conducted during the 3 monitoring campaigns (2017, 2018, 2019) and for each site, to help in
405 the interpretation of local air contamination data. These investigations were conducted over
406 a radius of 1 km around the air sampler installed at each site. The choice of the 1 km radius
407 was a compromise between technical feasibility (particularly in the wine-growing zone, where
408 the number of plots, often smaller than in field crops, is greater in a given area, implying a
409 greater number of farmers) and a surface area in agreement with atmospheric dispersion
410 patterns at the local scale. Such radius value was sufficient to find a correlation between
411 pesticide in precipitation and land use (Gryniewicz et al., 2001)). Atmospheric samples were
412 collected for a whole week during the spraying period (in average 27 weeks), this for 3 years
413 on the 7 sites concerned by the project, i.e. a total of 567 samples. Pesticide contents of each
414 sample were analyzed in an external accredited laboratory and allowed to quantify a.i.
415 concentration in the atmosphere.

416

417 **2.5. Evaluation of the predictive quality of I-Phy3**

418 Following Pierlot et al. (2017), two tests were carried out to compare outputs of I-Phy3
419 subindicators assessing the pesticide transfer to environmental compartments with
420 measured data (RC_{gw} ; RC_{sw} ; RC_{air} , see section 2.3.). First, a classical correlation test
421 between indicator outputs and measurements was carried out to calculate a correlation
422 coefficient r , and not the determination coefficient r^2 . The significance of the results by
423 calculating the p-value was also tested. Then, we ran a probability test consisting in comparing
424 the rank of indicator outputs and measurements through a contingency table. A similar ranking
425 means that the result of the indicator’ calculation appears to be correct while when the indicator
426 rank is lower than the rank of the measured value, it is considered as an underestimation and
427 when it is higher than the rank of the measured value, it is considered as an overestimation.
428 The probability considered in the test is the sum of correct and overestimation. Indeed, I-Phy
429 assesses a potential risk (which can occur or not depending on climate events for example),
430 so the positive result considered in this test are the sum of well-predicted events of transfer
431 and overestimation. (see a theoretical example in Table 6). Pierlot et al. (2017) set the rule that
432 an indicator is considered as acceptable when the probability is higher than 60% and the

433 correct estimation is higher than 40% to avoid considering an indicator whose results would
434 systematically predict a high risk regardless of the context. This general analysis was
435 completed by detailing the proportion of values in each class to assess the distribution of
436 values and to check that results are not only due to one class (e.g. the class no risk, no
437 pesticide in water).

438 For these analyses, regarding water contamination, the following measured variables available
439 in the EQUIPE project were used:

- 440 • frequency of exceedance of the threshold of the water quality standard of drinking
441 water: $0.1 \mu\text{g.L}^{-1}$ (fd0.1)

$$442 \text{fd0.1} = \frac{n_{ijk}^1}{n_{ijk}} \quad (\text{Equation 5})$$

443 with n_{ijk}^1 : number of measurements with concentration $> 0.1 \mu\text{g.L}^{-1}$ for active ingredient i on plot
444 j at sampling time k ; n_{ijk} : total number of measurements for active ingredient i on plot j and
445 sampling time k . The sampling was stopped when no a.i. was detected 3 consecutive weeks
446 and lasted one year maximum after the spraying date (Pierlot et al., 2017)

447

448

- 449 • cumulated flux of active ingredient in mg/ha (ftotal) during the measurement period

$$450 \text{ftotal} = \sum(f_{ijk}) \quad (\text{Equation 6})$$

451 with f_{ijk} : flux of active ingredient i on plot j and sampling time k ; $f_{ijk} = c_{ijk} \cdot w_{jk}$ with c_{ijk} : concentration
452 of active ingredient i on plot j and sampling time k ($\mu\text{g.L}^{-1}$) w_{jk} : water flux (drainage or runoff)
453 from plot j during sampling time k (L)

454

455

456

- 457 • weighted average concentration on the period in $\mu\text{g/L}$ (CMP)

$$458 \text{CMP} = \frac{\sum c_{ijk} \times w_{jk}}{\sum w_{jk}} \quad (\text{Equation 7})$$

459 with c_{ijk} : concentration of active ingredient i on plot j and sampling time k ($\mu\text{g.L}^{-1}$)

460 with c_{ijk} : concentration of active ingredient i on plot j and sampling time k ($\mu\text{g.L}^{-1}$)

461 with w_{jk} : water flux (drainage or runoff) from plot j during sampling time k (L)

462

463 Regarding the assessment of predictive quality for the atmospheric compartment, the
464 correlation between indicator outputs and a value calculated from measurements was
465 analyzed. Those from the REPP'AIR project were atmospheric concentrations in each site.
466 For a given site and year, the pesticide concentration in the sample was considered as
467 resulting from the volatilization and drift after spraying on fields from a buffer of 1km radius
468 around the sampler. Since temporal scales differed between spraying date (day), pesticide
469 concentration (week) and the indicator (year), it was not possible to compare the assessment
470 of volatilization of transfer by I-Phy calculated at field level and raw weekly concentrations. For
471 each a.i. of one site and one year, the weekly concentrations were plotted against the area
472 sprayed with a.i. during the week. The slope of the linear regression ion between concentration
473 and area was derived as a proxy of the measured volatilization risk. This means that for a given
474 sprayed area, the higher the slope, the higher the concentration expressing a higher
475 volatilization. This slope was compared to the mean of the indicator weighted by sprayed area

476 for each a.i. and for the sampling period. In this case, we worked on a limited number of points,
477 so that it was not possible to run a probability test.

478

479 **3. Results**

480 (Elliott et al., 2000; ERMES, 2017; Lindahl and Bockstaller, 2012; Melland et al., 2016; Roussel et al.,
481 2000; Vereecken, 2005; van der Werf and Zimmer, 1998)(ERMES, 2017; Koller et al., 2015)(Brown
482 and van Beinum, 2009; Buczko and Kuchenbuch, 2007; Carsel et al., 1986; Mickelson et al.,
483 2001; Pierlot et al., 2017; Strassemeyer and Gutsche, 2010; Trevisan et al., 2009; Wohlfart,
484 2008)
485 (Bahrouni et al., 2010; Bockstaller et al., 2017; Roussel et al., 2000; Thiollet-Scholtus and
486 Bockstaller, 2015; van der Werf and Zimmer, 1998; Woodrow et al., 1997)
487
488 (Centner, 2021; van der Werf and Zimmer, 1998)
489

490 **3.1. Examples of calculation**

491 I-Phy3 was calculated for 4 to 7 fields of 33 arable farms from the Champagne Crayeuse (East
492 of France) presenting diversified rotations with winter wheat, winter and spring barley, sugar
493 beet, potatoes, winter rapeseed, etc. for the harvest year of 2020. Figure 3 presents three
494 levels of results. From the top, at a first level, results for different level of intensity are shown
495 in for winter barley. They vary between 10and 2.5 for an intensive program with the herbicide
496 chlortoluron. This active ingredient presents a high risk for groundwater as shown at the
497 second level. Finally, explanation can be found at the third level. Chortoluron has very
498 unfavorable property regarding the GUS, the soil is sensitive to leaching and spraying period
499 in autumn are unfavourable because the soil becomes wet.

500

501 Tables at second and third levels on Figure 3 were directly taken from the Excel calculator
502 presenting the results of the final indicator and its sub indicators (see Figure 1). A continuous
503 colour code is used to provide information on the level of risk. Results of the sub indicators
504 (e.g. RCeso_{0.05}) are completed with the membership degree of each variable to the favourable
505 set (F), (see Supplementary 2). When this value was equal to 1, the variable is totally
506 favourable, i.e. it does not present a risk for the environment. This helps identify the ones which
507 influence the calculated risk and by this way can help users to identify levers to improve the
508 indicator and to reduce risk on the environment.

509 The example shows the ability of the indicator to differentiate crop management with different
510 level of intensity as well as risk level between active ingredients, and the possibility to explain
511 the results.

512

513 **3.2. Predictive quality of contamination risk subindicators**

514

515 **3.2.1. Predictive quality assessment for the water compartment**

516 Table 7 shows that the highest correlation between measured data in the fourth studied sites
517 (one for each transfer pathway) and the outputs of the risk of contamination indicator for ground
518 or surface water were obtained for the frequency of exceeding the threshold of the water quality

519 standard of drinking water (fd0.1), with value of correlation coefficient close to 0.50 (see
520 Supplementary Materials S12) and even more for $RC_{sw/d}$ at Geispitzen. ($r=0.66$). Such values of
521 r are close to those found by Pierlot et al; (2017) for indicator with the same degree of
522 complexity. These results are better for three sites or equal for one site (Jaillière runoff) than
523 those of the previous versions of I-Phy. The comparison between either the cumulated flux of
524 active ingredient in $mg \cdot ha^{-1}$ (ftotal) during the measurement period or the weighted average
525 concentration on the period in $\mu g \cdot L^{-1}$ (CMP), and outputs of the indicator yielded lower value of
526 coefficient between 0.08 and 0.35. For the site of Magneraud (leaching) and Geispitzen
527 (hortonian runoff), the new version yielded better value of correlation coefficients than the
528 previous one, which however remain at a lower level than for fd0.1, and much lower than 0.50.

529 The probability test reveals that the risk of contamination indicator for ground or surface water
530 yielded results meeting the criteria set for the test for fd0.1 and CMP for the fourth studied sites
531 (except for the site of Geispitzen for fd0.1). (Table 8). For ftotal, the only test meeting the
532 criteria set was for the site of Le Magneraud with a probability of 68% and correct estimation
533 of 49%. In comparison with the previous version of I-Phy, the new one obtains better results
534 than I-Phy2 for the three sites of La Jailliere runoff, Le Magneraud and Geispitzen for all the
535 tests, especially for the correct estimations. Compared to I-Phy1, I-Phy3 obtained better results
536 for the sites of Le Magneraud and Geispitzen for all the measured data whereas it surpassed
537 only for CMP for the site of La Jailliere runoff. From the analysis of the cases showing the
538 highest discrepancy (Table 9), it came out that 3 a.i. play a major role: epoxiconazole,
539 diflufenican and isoproturon explain 35 cases out of 50 .

540

541 **3.2.2. Predictive quality assessment for the air compartment**

542 Figure 4 shows a relatively clear correlation with a r of 0.73 between the slope coefficient of
543 the cumulative fluxes of a.i. and the means of risk of air contamination by volatilization ($RC_{air,vol}$),
544 this for only 12 a.i. for which there was enough data on the studied sites. Such a value is
545 satisfying regarding the elaboration degree of the indicator and when they are compared to the
546 coefficient for transfer to water. Nevertheless, unlike the risk of ground or surface water
547 contamination, the outputs of risk of air contamination were not compared directly to
548 contamination measurements at plot scale but to a calculated value derived from
549 measurements at a larger scale.

550

551

552 **4. Discussion**

553 **4.1 Originality of the I-Phy3 indicator**

554 I-Phy3 can be classified in the same class as I-Phy1 and I-Phy2 in the typology proposed by
555 Pierlot et al. (2017) classifying pesticide indicators assessing transfer risk to water in function
556 of their design. They are calculated with pesticide properties and use data, crop management
557 and field data (soil, slope, etc.). It does not only consist in a simple scoring of variables
558 according to expert opinion or an aggregation separating risk linked to pesticide properties,
559 and risk linked to soil and climate. Variables are integrated according to knowledge on
560 processes, with some calibration procedure for some subindicators. Furthermore, through the
561 decision rules, calculations seem to be more easy to grasp than indicators based on
562 quantitative equations like EPRIP2 (Trevisan et al., 2009), POCER (Vercruysse and Steurbaut,
563 2002) or SYNOPSIS (Strassemeyer and Gutsche, 2010). Regarding integration of toxicity
564 variables, those are aggregated in a qualitative way with transparent assumption in I-Phy3,

565 while in EPRIP2, SYNOPSIS or POCER, a risk ratio (concentration in the environment
566 compartment/concentration threshold for toxicological effect in this compartment) is used
567 resulting in a quantitative assessment. Exposure of living beings to pesticide is assessed with
568 more precision in POCER or in models used in Life Cycle Analysis (Gentil et al., 2020) taking
569 into account behavior of target living beings (e.g. for the ingestion exposure pathway) than in
570 the other indicators. Regarding I-Phy3, a sub indicator assessing exposure and effect may be
571 developed in the future by a separated decision tree and aggregated at the second level with
572 risk of contamination (RC) replacing the aggregation of RC with a toxicity variable (Figure 1).
573 A work is ongoing on pesticide effects on human health taking into account variables and
574 knowledge inputs from the POCER indicator (Vercruysse and Steurbaut, 2002) and the more
575 elaborated Browse model (Butler Ellis et al., 2017).

576
577 Aggregation procedure using fuzzy decision tree is also very original in comparison with other
578 indicators as pointed out by several authors (Feola et al., 2011; Keichinger et al., 2013; Maud
579 et al., 2001; Reus et al., 2002). This aggregation method presents different advantages like
580 the readability through linguistic rules, the possibility to cope with qualitative and quantitative
581 variables, the mitigation of threshold effect, section 2.2. For I-Phy3, the CONTRA method
582 (Bockstaller et al., 2017) was implemented to design fuzzy decision tree in order to enhance
583 transparency of the aggregation procedure. This confers a supplementary advantage to the
584 aggregation method, while aggregation is often criticized for a lack of
585 transparency. However, calculation of final result for a given decision tree may remain a “black
586 box” without additional information of intermediate calculation (Bockstaller et al., 2017). This
587 problem was partially solved as discussed further in section 4.4.
588

589 **4.2. Novelties of I-Phy3 compared to the previous versions**

590
591 The structure of I-Phy3 was totally changed compared to the initial version (van der Werf and
592 Zimmer, 1998) with the addition of a third level making it possible to deliver an assessment of
593 the contamination risk disconnected from the toxicity of the pesticide. Contamination of
594 environmental compartments is of major concern for many stakeholders working on water
595 quality management due to the current drinking water standards based on a concentration
596 threshold of $0.1 \mu\text{g.L}^{-1}$ independently from toxicity. This supplementary level may confer more
597 complexity to the indicator but this might not be a problem (see section 4.4.).
598

599 We tried to integrate more processes into the design of I-Phy3 to consolidate the scientific
600 basis. Nevertheless, the metamodelization approach derived from the mechanistic MACRO
601 model and implemented for the groundwater sub indicator in I-Phy2 was left. I-Phy2 version
602 did not yield satisfying results regarding its predictive quality for pesticide leaching (see Table
603 7). This discrepancy may be due to a parametrization of MACRO which did not deliver better
604 results for 6 out 7 parameter sets in the study of Pierlot et al. (2017). The new ground water
605 subindicator based on a simpler structure than this of I-Phy2 yielded better validation results
606 than I-Phy2 and slightly better results than I-Phy1.

607 The runoff surface water subindicator had already been improved in the second version
608 (Wohlfahrt, 2008). Besides the interception coefficient used for all indicators (i_c , see Equation
609 4), it is the only subindicator that entails the temporal dimension in an explicit way in the
610 availability variable (see Equation 2). Nevertheless, for most usage, this temporal variable
611 giving the time between spraying and variable is set to 3 days, which is a worst-case value like
612 in other indicators like Synops (Strassemeyer et al., 2017) and Eprip2 (Trevisan et al., 2009).
613 After all, it is still possible in the calculator to change the value and to make the indicator more
614 sensitive to the spraying date and the delay with the transfer event (*i.e.* significant rain). The
615 other subindicators were totally changed with additional information required, especially on
616 spraying conditions for spray drift to air and physical conditions of field margins. It was

617 assumed that information on spraying conditions and field margins characteristics are easily
618 accessible too.

619
620 Effect of tillage and pesticide incorporation were better integrated in the new version of I-Phy
621 by means of a much broader knowledge basis than for the previous version of I-Phy. Now the
622 effect was quantified more precisely than with a rough “expert value”, especially for effect of
623 tillage on runoff (meta-analysis of Elias, Wang, et Jacinthe (2018)). Such meta-analyses would
624 be useful to parametrize the effect of tillage on vertical transfer and the effect of pesticide
625 incorporation, for which some experimental results exist but remain fragmentary.

626
627 In I-Phy1, the effect of pesticide dose was assessed separately from the risk on environmental
628 compartment, the latter including the effect of crop interception. Like in I-Phy2, pesticide dose
629 and crop interception were combined since the amount that can be transferred from soil
630 surface to water is not the sprayed dose but depends on the interception by crop canopy.
631 Although a part of this amount intercepted by crop canopy can be washed off, we considered
632 like Rosenbaum et al. (2015) that in good practices conditions, pesticides are not sprayed just
633 before an important rainfall so that this fraction can be neglected in this approach. This
634 integration makes it possible to avoid giving systematically a favorable value to pesticide rate
635 with low application rate like it was the case in I-Phy1. As shown in Lopez et al. (2015) for
636 metsulfuron-methyl as well as in the ERMES monitoring program for nicosulfuron, sulfonyl-
637 urea herbicide sprayed at low rate (less 50 g/ha) are detected in groundwater and sometimes
638 at concentration exceeding quality water standards (Koller et al., 2015). With the new
639 calculation method, even for low a.i. application rates, unfavorable values for risk of
640 contamination subindicators may be found.

641 642 **4.3 Design of the indicator**

643
644 The indicator relies on an approach combining a qualitative (decision tree) with a quantitative
645 approach (fuzzy subsets) which present several advantages as pointed out previously. But the
646 outputs of the indicators are not expressed in quantitative physical or ecotoxicological values.
647 In particular, the indicator does not deliver quantitative information on contamination levels in
648 the environmental compartment, so that stakeholders have no information on the exceeding
649 of given standards like this for drinking water. This would require quantitative models which
650 are in most cases complicated to implement due to the type and amount of data required a
651 calibration procedure to carry out carefully to avoid false prediction as pointed out by Pierlot et
652 al. 2017 for MACRO. Another drawback of quantitative models is their reduced scope to one
653 or two environmental compartments. PestLCI 2.0 is an exception by covering the same
654 compartments as I-Phy3 (Dijkman et al., 2012) and providing percentages of emissions from
655 the initial rate in each compartment. However, it does not calculate concentration in the
656 environment and is only for about 100 active ingredients in comparison with about 500 for I-
657 Phy3.

658
659 In the assessment of the pesticide transfer pathways by many indicators, climate variables are
660 not directly included although variables like especially rainfall amount plays a significant role
661 in pesticide transfer (Baran et al., 2021). This would require additional data and may complexify
662 calculations. One way would be to integrate them in the leaching and runoff potential variable
663 in function of location and even of the year. This is possible manually in the calculator for dry
664 year; for example, it is easy to change the value into 0 (low potential). But adding an actual
665 value of the year may hide the effect of change of practice which is not the objective of the
666 indicator. Furthermore, intra annual effects of climate are taken into account by the period of
667 application according to the recommendation of Pierlot et al. (2017) for the groundwater
668 subindicator or by the availability variable (see Equation 5 for the surface subindicator). But in
669 case of transfer to air, climate is not included at all, neither for spray drift or volatilization. Wind

670 speed, a major driver for transfer to air (Lavin and Hageman, 2013), remains too difficult to get
671 for each treatment. Introducing such a variable would be an avenue for progress.

672

673 I-Phy3 like PestLCI 2.0 did not address contamination of soil by pesticides although several
674 recent studies revealed a “hidden reality” of pesticide in soils (Riedo et al., 2021; Silva et al.,
675 2019), especially for glyphosate (Silva et al., 2018), even in organic farming (Riedo et al.,
676 2021). Furthermore, these last authors found a negative relation between the amount of
677 pesticide residue in soil and microbial biomass and specifically the abundance of arbuscular
678 mycorrhizal fungi, a widespread group of beneficial plant symbionts likewise other parameters
679 of soil biological activities (Wolejko et al., 2020). The lack of a soil subindicator was due to
680 knowledge gaps which these recent studies tend to bridge. Besides the issue of soil pesticide
681 residues in terms of amount and concentration, another aspect concerns the temporal
682 dimension which should be included in future. Although a part of an active ingredient is
683 adsorbed in an irreversible way in the form of non-extractable bound residues (bounsten)
684 another non-negligible part may be released after several months (Suddaby et al., 2016).
685 Furthermore, the cumulative effect of repeated treatments should also be addressed.

686

687 Last, the contamination by metabolites released by the degradation of active ingredients
688 should also be assessed (Baran et al., 2021) as it has been pointed out by pioneer studies
689 (Dana W. Kolpin et al., 2000; D.W. Kolpin et al., 2000). But the integration of such an
690 assessment would probably complexify the indicator, exceeding an acceptable level. This
691 would require quantitative knowledge on the nature of the metabolite formed, the percentage
692 of a.i. transformed in this product, properties (KOC, DT50, etc.), etc., data that does not
693 currently exist in databases. except for recently marketed a.i. (Lopez et al., 2015). In any case,
694 some available information on metabolites was added indirectly in the database. For example,
695 in case when an active ingredient is rapidly degraded in its metabolites, (e.g. different form in
696 glyphosate acid, iodosulfuron-methyl in metsulfuron-metyl), properties of the metabolite are
697 attributed to a.i. For few pesticides like metazachlore, dimetolachlor (see Supplementary
698 Materials S1), selected values for DT50 appear to be too favourable while they present
699 metabolites susceptible to be transferred to water bodies (Reemtsma et al., 2013). In this case
700 we decided to attribute a more unfavorable value to the DT50 of this active ingredient.

701

702 **4.4 Predictive quality**

703 Datasets used to assess the predictive quality of the water contamination risk were
704 consequent. This was also the case for the air component in this study with several sites over
705 the country and a 3 years campaign. Nevertheless, as pointed out by Pierlot et al. (2017), the
706 effort should be pursued. Ideally, this work would require a broader combination of soil type,
707 slopes and climatic conditions, and much more new active ingredients.

708

709 The simplification performed in the design of I-Phy3 may explain the mixed results in the
710 validation test. Indeed, the compromise between accessibility of data and explanation of
711 mechanical processes leads us to ignore some variables like soil moisture, which is important
712 to explain transfers in pathway like agricultural drainage (Guimont et al., 2005) or wind speed
713 as explained above. It was considered that the additional cost for collecting usable data does
714 not compensate by loss of information for I-Phy3. Furthermore, these data are useful on a time
715 step of a few days when I-Phy3 estimates transfer risk at the scale of the growing season.

716

717 The results obtained for I-Phy3 were overall better than those obtained with the previous
718 versions. If the values of correlation coefficient are not always good, the results in the
719 probability test are satisfying, particularly for the frequency of exceeding the threshold of the
720 drinking water quality (fd0.1), particularly important for the stakeholders. The result of the test
721 combines acceptable and overestimation of risk. Thus, it shows the ability of the indicator to
722 assess a potential contamination which could or not be observed, depending on other factors
723 like climate. Unacceptable underestimations were observed for one site at la Jaillière runoff

724 dataset. It was showed that 3 a.i. (isoproturon, diflufenican and epoxiconazole) play a major
725 role in this underestimation. On this site, runoff was mostly to overland flow due to saturation
726 of the soil profile, a process which may be poorly covered by the indicator.

727
728

729 **4.5 Utilization**

730

731 The design of the I-Phy 3 indicator was not intended to be used directly by farmers themselves,
732 although this may be possible if farmers have time and support to interpret the results. It rather
733 targets advisers trained by scientists who may deliver interpreted results to feed the
734 recommendations to farmers in addition to technical advice. The statement of Box (1976): “all
735 models are wrong but some are useful”, may be applied to I-Phy3. In spite of its mixed results
736 regarding its predictive quality, it may be used beyond the simple results obtained by the
737 indicator. Indeed, it makes possible initiation of discussion on pesticide use strategies with
738 farmers integrating environmental aspects provided by the indicator and other aspects
739 (economic, management of weed and pest resistance, ...) not considered by the indicators.
740 Furthermore, if it does not give a precise value, it provides a positioning to some threshold,
741 like for instance to classify results in three classes (acceptable/mixed/unacceptable). But in
742 this case, we reintroduce a threshold effect that was avoided by the design method (see
743 section 2.2.). This is also the case with quantitative mechanistic models as PEARL (Tiktak et
744 al., 2012) which deliver continuous values but can be presented in class when they are mapped
745 for stakeholders.

746

747 For the council advice, as stated by Bockstaller et al. (2008), both aggregated indicators and
748 non-aggregated sub-indicators have to be used in parallel. The aggregated I-Phy indicator may
749 be associated with other sustainability indicators in a global assessment when there is a need
750 to reduce the number of indicators. In this case, non-aggregated subindicators should
751 complete and explain the global value in the analysis. The aggregated I-Phy indicator may also
752 serve to rank pesticides when all the environmental issues are considered by stakeholders
753 while a non-aggregated subindicators, especially risk of contamination indicators may be
754 implemented by stakeholders dealing with one environmental compartment (e.g.
755 groundwater).

756

757 Currently the indicator is calculated with the help of an Excel sheet calculator, which facilitates
758 its implementation because the software is a basic software on most of the users' offices. The
759 tool is transparent for the user, who can see all the decision rules. The problem is that it may
760 also lead to misuse because the user can potentially and accidentally change the decisions
761 rules. This can be solved by protection of some cells. However, misuse also occurs in copying
762 and pasting lines, deleting formulas in the cell. To mitigate the problem, we can imagine a
763 further online-version of the tool with data fields to be completed and calculations of the
764 different sub-indicators provided. In any case, the tool should remain transparent and not only
765 provide calculation results but also the intermediate values to interpret them and identify the
766 variable(s) which plays the major role in the determination of the risk.

767

768 Last but not least, the I-Phy indicator is designed to predict the risk of pesticide transfer at field
769 level. Some of the stakeholders need to assess the risk of transfer at a higher level like a
770 watershed or an administrative region (i.e. the managers of the Water Agencies in France or
771 the advisers in charge of water catchments). Some previous research work ((Wohlfahrt et al.,
772 2010) shows that the contribution of the different plots to the watershed depends on the size
773 of the plot in comparison to the size of the watershed and the distance and the connectivity
774 between the plot and the hydrological network. To simplify the use of the indicator at watershed
775 scale and because they have no precise information about the real water flows in the
776 watershed, advisers consider only the size of the plot in comparison to the size of the
777 watershed. Another approach is to map the distribution of the indicator values set in small

778 classes like a 3 levels scale as traffic lights without any spatial aggregation, as discussed at
779 the beginning of this section.
780

781 **5. Conclusion**

782 This new version of the I-Phy indicator and its subindicators provides major changes in
783 comparison with previous versions, to better integrate processes of transfer of the pesticides
784 to the environmental compartment. The separation between contamination and toxicity, as well
785 as the transfer pathways are some examples to meet requirements of potential users like
786 adviser or stakeholders. For the air compartment, a spray drift subindicator was added but like
787 previous versions, whereas soil is still not addressed by this indicator, partially due to
788 knowledge gaps. With the increasing focus on this compartment in publications, this gap is
789 about to be filled and an addition of a new subindicator will be possible. While the effect of
790 degradation products of the a.i. is covered by I-Phy3 in an indirect way, their risks should be
791 covered in an explicit way to meet social concern for this issue. Last, the effort to assess the
792 predictive quality of the indicator should be pursued and should be completed by a feasibility
793 and utility test among end-users. This new version should still be confronted with other
794 datasets including recent active ingredients with the aim of improving the predictive quality.
795

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1035 Table 1: Snapshot of CONTRA “decision tree” tab (Bockstaller et al., 2017), showing the
 1036 decision tree design of the subindicator, pesticide leaching risk of contamination (RC_{gw_{lea}}). The
 1037 groundwater ubiquity index (GUS), the application period (Appli), the leaching potential of soil
 1038 (LixPot) and the solubility of the active ingredient in water (Solu) were aggregated with
 1039 respectively a weight of 40% for GUS and 20% for the three other variables, giving the
 1040 calibrated RC_{gw_{lea}} expressed between 0 (high risk) and 10 (low risk). Some modifications were
 1041 given to the decision rules yielding the final RC_{gw_{lea}} according to the explanations given in the
 1042 last column. Input variables belong either to the favourable set (F) or to unfavourable set (U),
 1043 (see Table S3).

1044

GUS	Appli	LeaPot	Solu	Calibrated RC _{gw_{lea}}	Correction (absolute value)	Final RC _{gw_{lea}}	Explanation on correction
F	F	F	F	10,0		10,0	
F	F	F	D	8,0	1	9,0	if GUS is favourable, effect of solubility is reduced
F	F	D	F	8,0		8,0	
F	F	D	D	6,0	1	7,0	if GUS is favourable, effect of solubility is reduced
F	D	F	F	8,0		8,0	
F	D	F	D	6,0	0,5	6,5	if GUS is favourable, effect of solubility is reduced
F	D	D	F	6,0		6,0	
F	D	D	D	4,0		4,0	
D	F	F	F	6,0	-0,5	5,5	if GUS is unfavourable, risk is increased to avoid a similar results with previous lines
D	F	F	D	4,0	0,5	4,5	if praying period is favourable (dry soil), risk is reduced
D	F	D	F	4,0	0,5	4,5	if praying period is favourable (dry soil), risk is reduced
D	F	D	D	2,0	0,5	2,5	if praying period is favourable (dry soil), risk is reduced
D	D	F	F	4,0		4,0	
D	D	F	D	2,0		2,0	
D	D	D	F	2,0		2,0	
D	D	D	D	0,0		0,0	

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1047 Table 2: Decision tree of the subindicator, pesticide runoff risk of contamination (RC_{sw_{r/d}}) in
 1048 case for runoff. RuPot is the runoff potential and Avai is the availability of the pesticide (see
 1049 Equation 5). Input variables belong either to the favourable set (F) or to unfavourable set (U),
 1050 (see Table S3).

1051

RuPot	Avai	RC _{sw_{r/d}}
F	F	10
F	U	5.5
U	F	6.7
U	U	0

1052

1053 Table 3: Snapshot of CONTRA “decision tree” tab (Bockstaller et al., 2017), showing the
 1054 decision tree design of the subindicator, risk of air contamination by drift (RC_{air_d}). The type of
 1055 sprayer (Sprayer), the speed sprayer (Speed), the sprayer height (Height), the use of antidrift

1056 nozzle (Nuzzle) and the air pressure (Pressure) were aggregated with the same weight of 20%
 1057 for all variables, giving the calibrated $RC_{air,dr}$, expressed between 0 (high risk) and 10 (low risk).
 1058 Some modifications were given to the decision rules yielding the final $RC_{air,dr}$ according to the
 1059 explanations given in the last column. Input variables belong either to the favourable set (F) or
 1060 to unfavourable set (U), (see Table S3).

Sprayer	Speed	Height	Nuzzle	Pressure	Calibrated $RC_{air,dr}$	Correction (absolute value)	Final $RC_{air,dr}$	Explanation on correction
F	F	F	F	F	10	1	9,0	Drift is possible even if all variables are favourable
F	F	F	F	U	8		8,0	
F	F	F	U	F	8		8,0	
F	F	F	U	U	6		6,0	
F	F	U	F	F	8		8,0	
F	F	U	F	D	6		6,0	
F	F	U	U	F	6		6,0	
F	F	U	U	U	4		4,0	
F	U	F	F	F	8	0.5	7.5	If sprayer speed is unfavourable then an additional negative effect
F	U	F	F	U	6	0.5	5.5	If sprayer speed is unfavourable then an additional negative effect
F	U	F	U	F	6	0.5	5.5	If sprayer speed is unfavourable then an additional negative effect
F	U	F	U	U	4	0.5	3.5	If sprayer speed is unfavourable then an additional negative effect
F	U	U	F	F	6	0.5	5.5	If sprayer speed is unfavourable then an additional negative effect
F	U	U	F	U	4	0.5	3.5	If sprayer speed is unfavourable then an additional negative effect
F	U	U	U	F	4	0.5	3.5	If sprayer speed is unfavourable then an additional negative effect
F	U	U	U	U	2	0.5	1.5	If sprayer speed is unfavourable then an additional negative effect
U	F	F	F	F	8	8	0	If sprayer type defavourable, no possibility to reduce drift
U	F	F	F	U	6	6	0	If sprayer type defavourable, no possibility to reduce drift
U	F	F	U	F	6	6	0	If sprayer type defavourable, no possibility to reduce drift
U	F	F	U	U	4	4	0	If sprayer type defavourable, no possibility to reduce drift
U	F	U	F	F	6	6	0	If sprayer type defavourable, no possibility to reduce drift
U	F	U	F	U	4	4	0	If sprayer type defavourable, no possibility to reduce drift
U	F	U	U	F	4	4	0	If sprayer type defavourable, no possibility to reduce drift
U	F	U	U	U	2	2	0	If sprayer type defavourable, no possibility to reduce drift
U	U	F	F	F	6	6	0	If sprayer type defavourable, no possibility to reduce drift
U	U	F	F	U	4	4	0	If sprayer type defavourable, no possibility to reduce drift
U	U	F	U	F	4	4	0	If sprayer type defavourable, no possibility to reduce drift
U	U	F	U	U	2	2	0	If sprayer type defavourable, no possibility to reduce drift
U	U	U	F	F	4	4	0	If sprayer type defavourable, no possibility to reduce drift
U	U	U	F	U	2	2	0	If sprayer type defavourable, no possibility to reduce drift
U	U	U	U	F	2	2	0	If sprayer type defavourable, no possibility to reduce drift
U	U	U	U	U	0		0	If sprayer type defavourable, no possibility to reduce drift

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1064 Table 4: Snapshot of CONTRA “decision tree” tab (Bockstaller et al., 2017), showing the
 1065 decision tree design of the subindicator, pesticide risk for surface water (I-Phy_{sw}). The
 1066 contamination risk for surface water through drainage or runoff ($RC_{sw,dr}$), the contamination
 1067 risk for surface water through drift ($RC_{sw,dr}$), and the toxicity variable for aquatic organisms
 1068 (TOX) were aggregated with respectively a weight of 33%, giving the calibrated I-Phy_{sw}
 1069 expressed between 0 (high risk) and 10 (low risk). Some modifications are given to the
 1070 decision rules yielding the final I-Phy_{sw} according to the explanations given in the last column.
 1071 Input variables belong either to the favourable set (F) or to unfavourable set (U), (see Table
 1072 S3).

RCsw _{gl}	RCsw _{gr}	Tox	Calibrated I-Phy _{sw}	Correction (absolute value)	Final I-Phy _{sw}	Explanation on correction
F	F	F	10		10	
F	F	U	6		6	
F	U	F	7		7	
F	U	U	3	-1	2	Transfer for one pathway and toxicity unfavourable: decrease of 1 point to highlight the unacceptable situation for stakeholder
U	F	F	7		7	
U	F	U	3	-1	2	Transfer for one pathway and toxicity unfavourable: decrease of 1 point to highlight the unacceptable situation for stakeholder
U	U	F	4		4	
U	U	U	0		0	

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1075 Table 5: Snapshot of CONTRA “decision tree” tab (Bockstaller et al., 2017), showing the
 1076 decision tree design calculating the final I-Phy indicator. The subindicators assessing the risk
 1077 for three environmental compartments, pesticide risk respectively for groundwater (I-Phy_{gw}), for
 1078 surface water (I-Phy_{sw}) and for air (I-Phy_{air}) were aggregated with the same weight of 33%, giving
 1079 the calibrated I-Phy expressed between 0 (high risk) and 10 (low risk). Some modifications
 1080 were given to the decision rules yielding the final I-Phy according to the explanations given in
 1081 the last column. Input variables belong either to the favourable set (F) or to unfavourable set
 1082 (U), (see Table S3). Membership functions are sinusoidal.

I-Phy _{gw}	I-Phy _{sw}	I-Phy _{air}	Calibrated I-Phy	Correction (absolute value)	Final I-Phy	Explanation on correction
F	F	F	10		10	
F	F	U	6.7	-0.7	6	To set at 6
F	U	F	6.7	-0;7	6	To set at 6
F	U	U	3.3	-1.3	2	To set at 2
U	F	F	6.7	-0.7	6	To set at 6
U	F	U	3.3	-1.3	2	To set at 2
U	U	F	3.3	-1.3	2	To set at 2
U	U	U	0		0	

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1084 Table 6: theoretical example of probability test comparing classes of an indicator (in colum,
 1085 class 5 shows a higher value effect than 1) with a measurement (increasing values show a
 1086 higher effect). The probability is the sum of correct estimation (cases colored in green) and
 1087 overestimation (cases colored in blue). The cases colored in brown are considered as
 1088 underestimation.

Result of Indicator (in class)	Result of measured data (in class)				
	From 0 to 20	From 20 to 40	From 40 to 60	From 60 to 80	Fomr 80 to 100
From 8 to 0	20	8	8	9	2
From 6 to 8	16	5	11	11	35

From 4 to 6	3	1	12	22	3
From 2 to 4	23	0	0	1	5
From 0 to 2	4	0	1	0	2

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1092 Table 7: correlation test between the measured data of the 3 sites and 4 transfer pathway
 1093 and the concerned subindicator RC for the 3 versions of I-Phy. The results are in bold when
 1094 subindicators of I-Phy3 performs better than in the previous versions.

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Component of Phy	Site	Transfer pathway	fd0.1			ftotal			CMP		
			I-Phy3	I-Phy 1	I-Phy 2	I-Phy3	I-Phy 1	I-Phy 2	I-Phy3	I-Phy 1	I-Phy 2
RC _{pa-1ea}	La Jaillière	Drainage	0.48	0.36	0.24	0.35	0.49	0.19	0.35	0.41	0.13
RC _{ca-n/d}	La Jaillière	Saturation runoff	0.49	0.44	0.49	0.27	0.26	0.28	0.32	0.38	0.31
RC _{ca-n/d}	Geispitzen	Hortonien runoff	0.66	0.31	0.32	0.15	-0.15	-0.04	0.35	0.21	0.34
RC _{pa-1ea}	Le Magneraud	Leaching	0.46	0.3	-0.17	0.17	0.05	-0.11	0.08	-0.01	-0.09

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1099 Table 8: probability test (see Figure 2) for the 3 sites and 4 transfer pathways for each
 1100 concerned subindicator RC (see Figure 3), comparing to the measured transfer of active
 1101 ingredient In blue the probability is over 70%, in yellow, the probability is between 50% and
 1102 70% and in red, between 40% et 50%

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Component of I-Phy 3	site	fd 0.1				ftotal				CMP			
		probability	correct	over estimation	under estimation	probability	correct	over estimation	under estimation	probability	correct	over estimation	under estimation
Jaillère ruisselement	I-Phy 3 esu (not e)	53%	43%	10%	47%	52%	39%	13%	48%	53%	41%	12%	47%
Jaillère drainage	I-Phy 3 Reso (not e)	74%	44%	30%	26%	57%	38%	19%	43%	68%	45%	23%	32%
Magneraud	I-Phy 3 Reso (not e)	81%	56%	26%	19%	68%	49%	18%	32%	79%	55%	24%	21%
Geispitzen	I-Phy 3 Resu (not e)	73%	28%	45%	28%	80%	28%	53%	20%	73%	40%	33%	28%

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1134 Table 9: detailed probability test (see Figure 2) for the site of La Jaillière runoff for the frequency
 1135 of exceedance of the threshold of $0,1\mu\text{g.L}^{-1}$.

Class	Fd 0.1 (%)					n	%
	20	40	60	80	100		
1	80	8	8	9	14	Correct	90 43%
2	16	8	11	11	27	overestimation	20 10%
3	3	1	2	1	3	underestimation	97 47%
4	0	0	0	0	5	Total	207
5	0	0	0	0	0		

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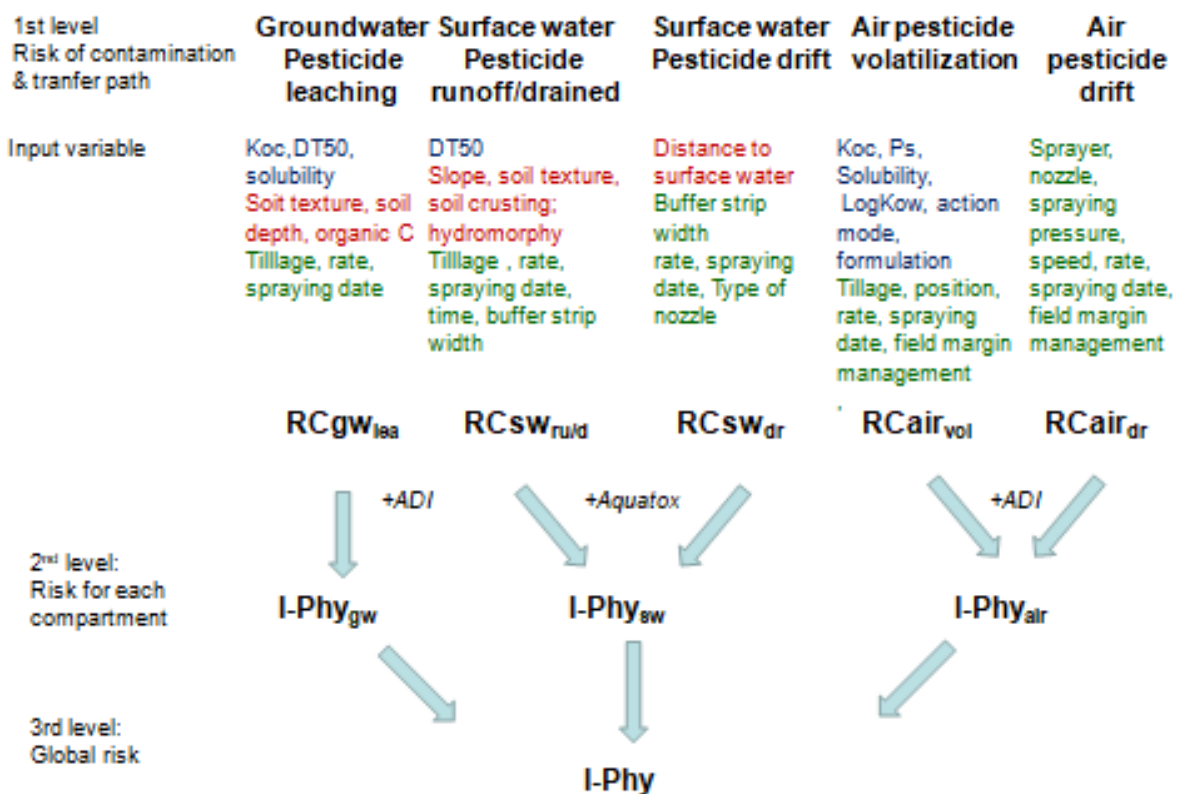
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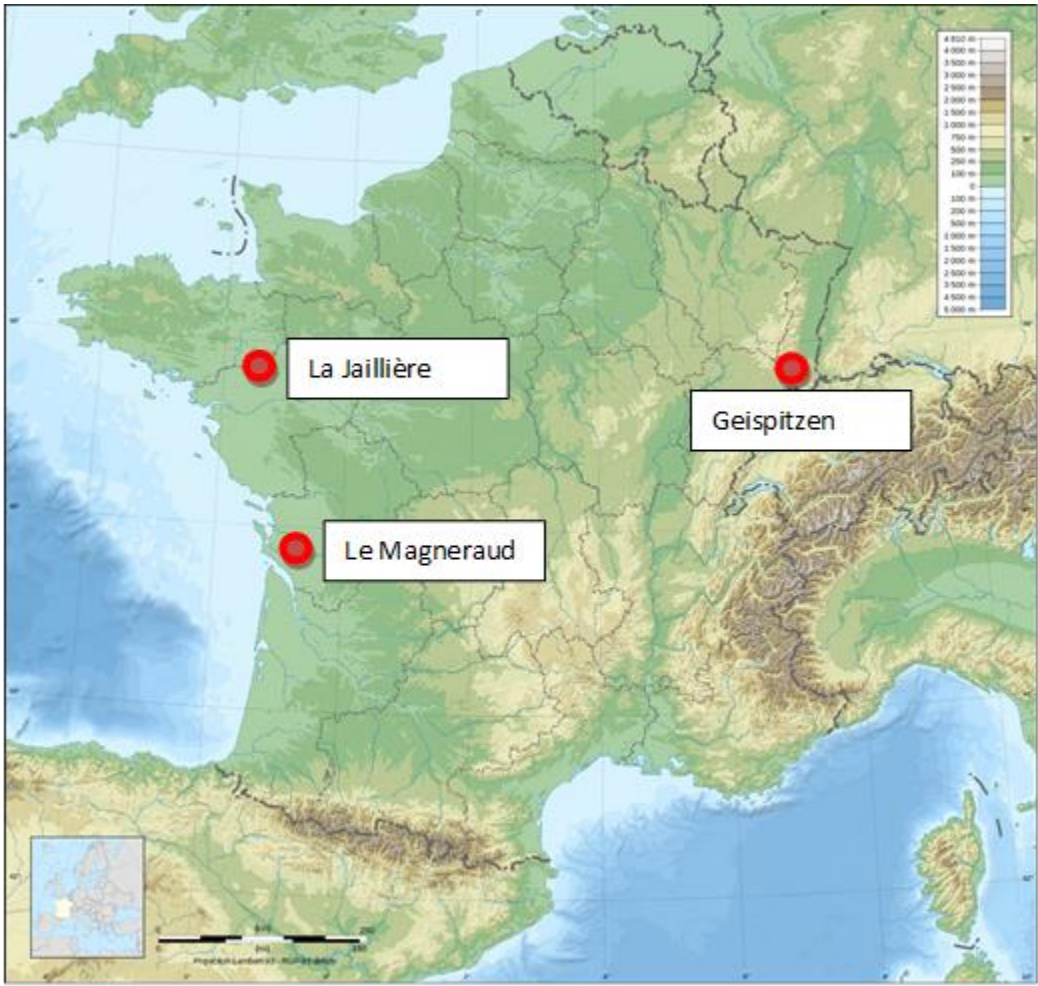




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1146 Figure 1: Overview of the calculation of I-Phy3 with three levels of aggregation and main input
 1147 variables, in blue pesticide properties, in red soil and topographic variables, in green:
 1148 management variable (KOC: adsorption coefficient, DT50: half-life, Solubility: solubility in
 1149 water, Ps: vapor pressure, LogKow: logarithm of the octanoal-water coefficient, time: time
 1150 between spraying and runoff event). For each aggregation, a fuzzy decision tree (see Figure
 1151 S2) was implemented with information on fuzzy subsets linked to each variable given in Table
 1152 S2.

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 1155 Figure 2: Repartitions of the sites used to assess predictive quality. With red bullet: sites for
 1156 water quality and with blue bullets: sites for air quality

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Year	Crop	Active ingredient	Corrected rate (g/ha)	% treated area	I-Phy	I-Phy _{gw}	RC _{gw}	I-Phy _{sw}	RC _{sw-rd}	RC _{sw-dr}	I-Phy _{air}	RC _{air-vol}	RC _{air-dr}
2020	Spring barley	2,4-MCPA	150	100	4.6	4.3	3.6	6.9	6.6	10.0	4.7	6.1	4.9
2020	Spring barley	clopyralid	15	100	7.8	6.5	5.6	9.0	8.6	10.0	8.2	10.0	6.9
2020	Spring barley	fluroxypyr	30	100	9.0	8.8	7.0	9.1	8.5	10.0	8.8	10.0	5.8
2020	Spring barley	éthéphon	24	100	8.2	9.0	10.0	8.4	8.6	10.0	7.2	9.2	5.8
2020	Spring barley	prothioconazole	13	100	8.3	9.0	10.0	8.2	10.0	10.0	7.7	10.0	6.9
2020	Spring barley	fluoxastrobine	6	100	7.1	8.3	10.0	7.3	9.5	10.0	7.4	10.0	7.4

RC _{gw} (weighted by treated area)	Applicati on GUS	Potential of leaching (day num ber)	Solubility
3.6	0.00	0.55	0.10
5.6	0.00	0.56	0.10
7.0	0.31	0.56	0.10
10.0	1.00	1.00	0.10
10.0	1.00	1.00	1.00
10.0	0.98	1.00	0.10

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Crop	Crop mnagement	Spraying programm*	I-Phy
Winter barley	Intensive (without chlortoluron)	3H, 2F, 11, 3R	2.5
Winter barley	Intensive (without chlortoluron)	5H, 5F, 21, 3R	4.1
Winter barley	Integrated	4H, 3F, 01, 1R	6.5
Winter barley	Organic	0H, 0F, 01, 0R	10.0

* Number of herbicides (H) fungicides (F), insecticides (I), growth regulator (R)

Active ingredient	Rate (g/ha)	I-Phy	I-Phy _{gw}	RC _{gw}	I-Phy _{sw}	RC _{sw-rd}	RC _{sw-dr}	I-Phy _{air}	RC _{air-vol}	RC _{air-dr}
flufenacet	200	4.2	5.1	5.2	3.7	4.6	10.0	6.2	10.0	7.4
diflufenican	100	6.5	8.9	8.2	3.8	4.7	10.0	8.5	10.0	9.4
chlortoluron	1250	3.5	3.5	2.1	2.9	2.7	9.7	5.7	9.9	8.5
esfenvalerate	6	6.8	8.5	10.0	5.4	7.6	10.0	8.2	10.0	7.9
cyprodinil	143	6.3	8.6	9.5	5.4	4.7	10.0	6.6	9.1	8.3
ethephon	143	6.3	8.0	8.1	6.8	5.1	10.0	6.3	8.2	9.3
chlormequat chlorure	285	5.3	5.5	4.7	6.9	4.5	10.0	6.9	10.0	9.3
prothioconazole	39	8.2	9.0	10.0	8.2	10.0	10.0	7.4	10.0	10.0
benzovindiflupyr	20	7.8	10.0	10.0	4.9	6.5	10.0	9.3	10.0	5.5
ethephon	54	7.4	9.0	9.9	7.1	6.2	10.0	6.9	8.9	5.8

Indicator	Membership value: 1 if favourable, 0 if unfavourable			
RC _{gw}	GUS	Spraying period	Leaching potential	Solubility
5.2	0.22	0.00	0.10	1.00
8.2	1.00	0.00	0.10	1.00
2.1	0.00	0.00	0.10	1.00
10.0	1.00	0.00	0.10	1.00
9.5	1.00	0.63	0.10	1.00
8.1	1.00	0.63	0.10	0.00
4.7	0.13	0.63	0.10	0.00
10.0	1.00	0.88	0.10	1.00
10.0	1.00	0.88	0.10	1.00
9.9	1.00	0.88	0.10	0.00

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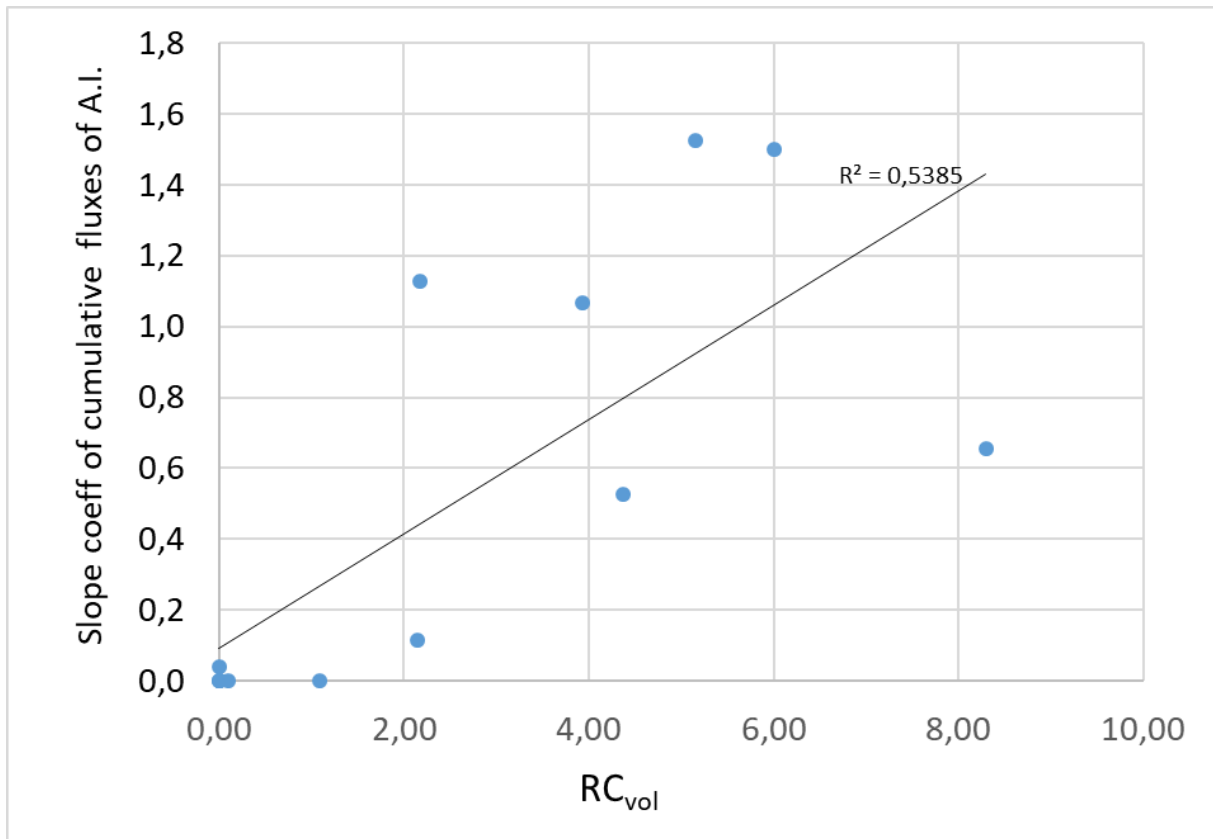
1166 Figure 3: the detail for one subindicator RC_{Ceso}. The membership degrees of the input variables
 1167 show to which extent the variable is unfavourable (close to 0) or favourable (close to 1) and
 1168 plays a role in the determination of the risk.

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1174 Figure 4: correlation between the slope coefficient of the cumulative fluxes of a.i. used as a
 1175 proxy of volatilization risk (see SM XX) and the means of RC_{vol} for 12 a.i.

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